

# **Observations Derived from the Characterization of Monolithic Fuel Plates Irradiated as Part of the RERTR-6 Experiment**

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# OBSERVATIONS DERIVED FROM THE CHARACTERIZATION OF MONOLITHIC FUEL PLATES IRRADIATED AS PART OF THE RERTR-6 EXPERIMENT

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## ABSTRACT

Evaluation of the PIE results of the monolithic plates that were irradiated as part of the RERTR-6 experiment has continued. Specifically, comparisons have been made between the microstructures of the fuel plates before and after irradiation. Using the results from the rigorous characterization that was performed on the as-fabricated plates using scanning electron microscopy, it is possible to improve understanding of how monolithic fuel plates perform when they are irradiated. This paper will discuss the changes that occur, if any, in the microstructure of a monolithic fuel plate that is fabricated using techniques like what were employed for fabricating RERTR-6 fuel plates. In addition, the performance of fuel/cladding interaction layers that were present in the fuel plates due to the fabrication process will be discussed, particularly in the context of swelling of these layers and how these layers exhibit different behaviors depending on whether the fuel alloy in the fuel plate is U-7Mo or U-10Mo.

## 1. Introduction

Monolithic fuel plates are being developed as an LEU fuel for application in research reactors [1]. To fabricate these fuels, three techniques have been evaluated: friction bonding (FB), hot isostatic pressing (HIP), and transient liquid phase bonding (TLPB) [2]. To better understand observations from the RERTR-6 experiment, which tested FB monolithic plates irradiated at moderate powers (surface heat flux  $\sim 140\text{--}175\text{ W/cm}^2$ ), at high temperatures (centerline temperature at BOL  $\sim 116\text{--}180^\circ\text{C}$ ) and at moderate burnups ( $\sim 50\%$  LEU) [1], as-fabricated FB monolithic plates, which were produced as archives were characterized using SEM/EDS/WDS analysis. Significant interaction was observed between the U-7Mo alloy and the Al-6061 cladding in these plates, due to their exposure to relatively high temperatures during the flattening ( $385^\circ\text{C}$ , 3-4 min.) and homogenization anneal ( $500^\circ\text{C}$ , 30 min.) steps, and lesser interaction was observed for the as-fabricated U-10Mo plates. For the as-irradiated fuel plates, optical metallography was employed to determine the fuel plate microstructures. By comparing the results of the as-fabricated fuel plate characterization to that of the as-irradiated fuel plates, one can make observations about the irradiation performance of monolithic RERTR fuel plates. In particular, the effects, if any, of having significant fuel/cladding interaction zones at the fuel/cladding interface and Al-rich phases present all the way to the center of a fuel foil can be better understood.

## 2. Experimental

Cross sections of fuel plates, which were fabricated exactly like those that went into the RERTR-6 reactor experiment, were characterized using a ZEISS 960A scanning electron microscope (SEM) equipped with energy-dispersive and wavelength-dispersive spectrometers (EDS/WDS). The as-fabricated FB U-7Mo monolithic fuel plates that were characterized included N1F070 and N1F080, and the as-irradiated FB U-7Mo fuel plates that were characterized were N1F030 and N1F090. For the FB U-10 Mo plates, samples L1F080, L1F110, and L2F010 were the as-fabricated samples that were characterized, and the as-irradiated samples included L1F040, L2F030, and L1F100. SEM images for the various as-fabricated samples were compared to

optical images for the as-irradiated plates, which were produced using a metallograph located in the Hot Fuel Examination Facility. Of particular interest were any changes that had developed in the fuel plate microstructure due to the irradiation process.

### 3. Results

#### 3.1 U-7Mo Plates

##### 3.1.1 N1F030

In Figure 1, representative microstructures for the as-irradiated plate N1F030 are presented. A uniform interaction layer is present at the fuel/cladding interface. Figure 2 shows that during the fabrication process a fuel/cladding interaction zone can form. Based on the narrow thickness observed in plate N1F030, along with the layer morphology, it is not clear that this layer formed due to fabrication. This cross-section may have been taken at an area where appreciable as-fabricated reaction was not present. Figure 3 shows how U-7Mo plates can have areas of appreciable reaction, but also areas of negligible reaction. When layers form during fabrication, they do not look like the narrow, uniform layer depicted in Figure 1. This suggests the layer formed during irradiation.

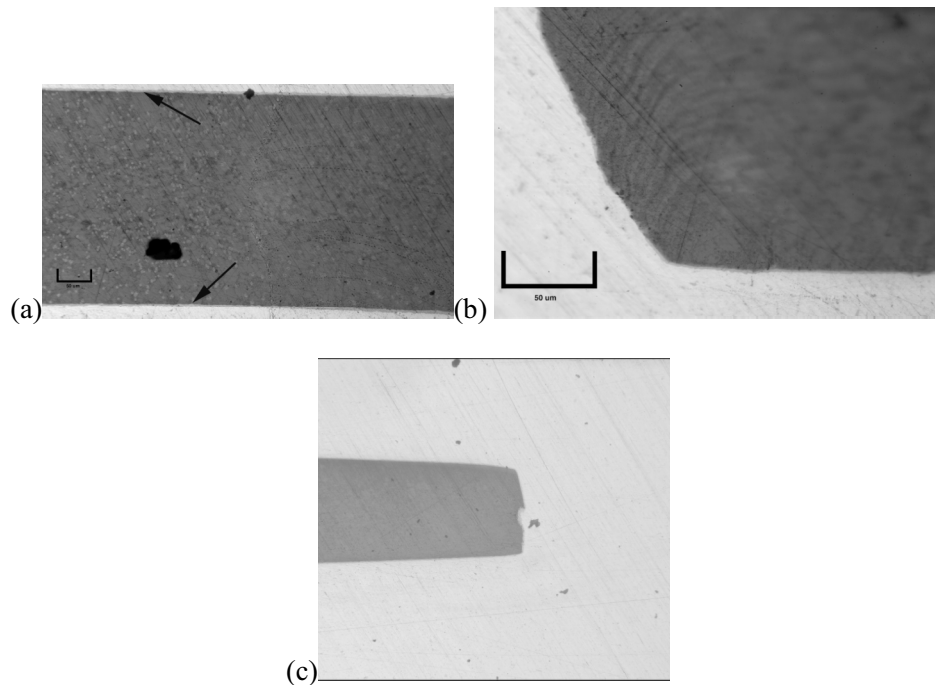


Figure 1. Optical images of irradiated fuel plate N1F030 that show the presence of a uniform reaction layer at most locations along the fuel/cladding interface.

The irradiation process also may have resulted in the development of porosity in the interaction layer. The porosity that is present in some regions of the interaction layer is shown in Figure 4. This porosity was not present after fabrication, based on the SEM analyses of fuel samples that were fabricated in the same manner as N1F030. Before attributing the creation of these pores to the irradiation process, one must rule out sample pull out during polishing as a possible cause of the porosity. As a result, a second transverse cross-section was generated for this sample, to see if more porosity could be observed. As shown in Figure 4d, appreciable porosity was also found in local areas of the interaction zone for this sample. The morphology of these pores were very

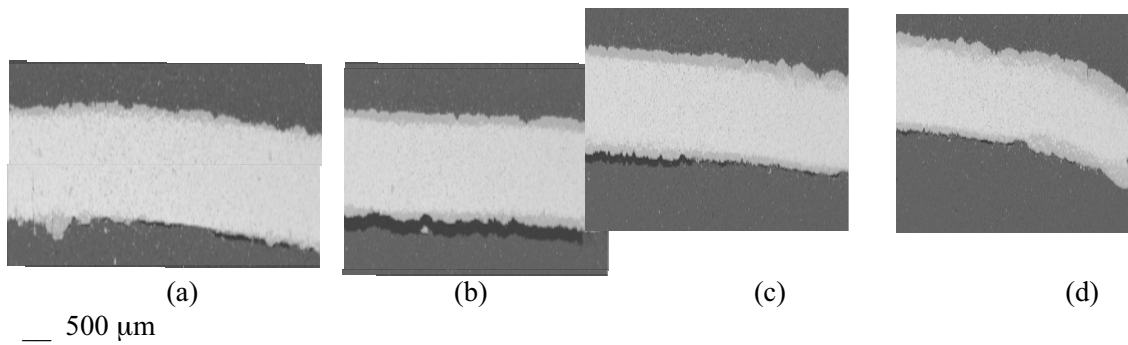


Figure 2. Backscattered electron images along a transverse cross section of as-fabricated fuel plate N1F070 that show the presence of an interaction zone at most locations along the fuel/cladding interface.

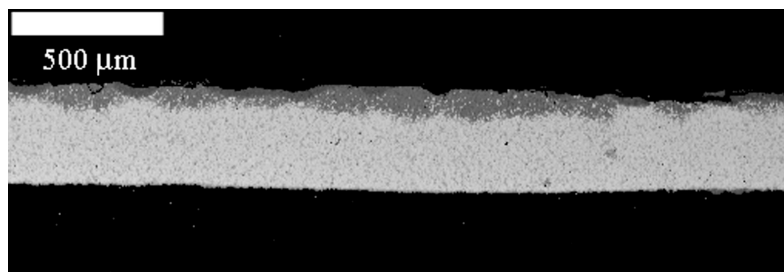


Figure 3. Backscattered electron images of a transverse cross-section taken from as-fabricated plate N1F080.

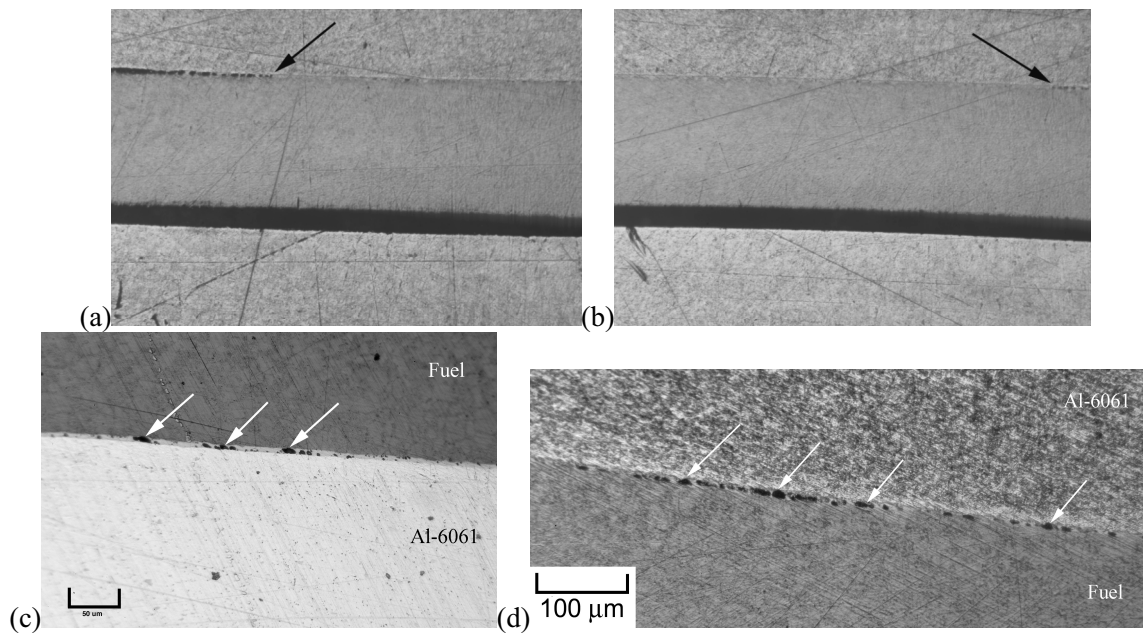


Figure 4. Optical images (a,b) along a transverse cross-section of irradiated fuel plate N1F030. The arrows indicate where the interaction layer transitions to an area where porosity has developed. The darkest contrast region is where a gap formed between the foil and cladding during sample preparation. (c) is a higher magnification optical image showing the porosity (arrows) present in the interaction layer. (d) shows porosity that is present in the interaction layer for a second transverse cross-section taken from plate N1F030.

similar to those observed in dispersion fuels, which are created during irradiation [3]. When sample pullout occurs in samples, typically large sections of the reaction layer are removed, not very small areas that leave void space with rounded, smooth edges. If sample pull out was such a problem when preparing these samples, then it should be much more common since all the irradiated samples are prepared in the same manner, and this is not the case. Therefore, it appears likely that the porosity in sample N1F030 is a result of the irradiation process.

Besides the interaction layer, another type of phase that was present in these fuel plates, before they were irradiated, was an Al-containing phase that could be found throughout the foil. An optical image showing this phase in the irradiated plate N1F030 is presented in Figure 5. Not much change (e.g., development of porosity) seems to have occurred in the two-phase foil microstructure during irradiation.

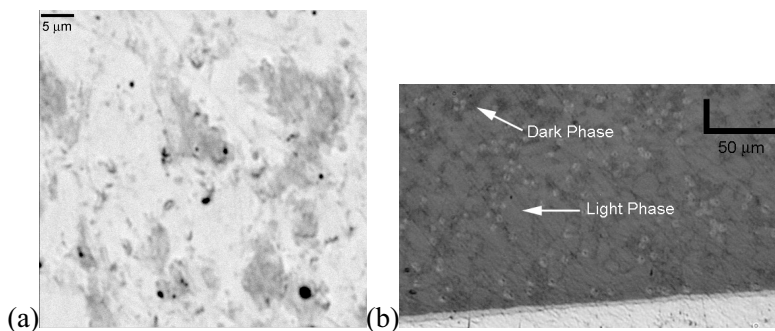


Figure 5. (a) Backscattered electron image of the two-phase foil microstructure in the as-fabricated fuel plate N1F080 and (b) an optical image of the two-phase foil microstructure for the as-irradiated plate N1F030.

### 3.1.2 N1F090

The as-irradiated fuel plate N1F090 exhibited significantly more fuel/cladding interaction than did N1F030 (see Figure 6). The interaction zone formed as a result of the fabrication process. Based on Figures 3 and 7, which show the great variations in the thickness of the fuel/cladding interaction zones for as-fabricated samples N1F080 and N1F070, respectively, it can be seen that there can be great variation in the thickness of the interaction zone depending on the location at the fuel/cladding interface. In Figure 6, there are local regions where it looks like there is negligible interaction along the fuel/cladding interface. This means it is possible that if another transverse cross section was characterized from N1F030 more, or less, fuel/cladding interaction may be observed. Thus, there may not be such a great difference in interaction zone size between samples N1F030 and N1F090. The overall attributes of the interaction layer observed in N1F090 agree with what has been observed when characterizing the as-fabricated monolithic plates.

## 3.2 U-10Mo Plates

### 3.2.1 L1F040

Optical images that were taken of a transverse cross-section from fuel plate L1F040 are presented in Figure 8. The layer that is present at the fuel/cladding interface is narrow and uniform. These images look similar to SEM micrographs that were taken for as-fabricated samples taken from U-10Mo fuel plates (L2F010, L1F110, and L1F080) in that not much of an apparent reaction layer is present at the fuel/cladding interface. This is contrary to the U-7Mo as-fabricated plates discussed earlier, which typically had more significant reaction layers at the interface. Figures 9c-9e show some images from the as-fabricated plates where no fuel/cladding interaction layer is observed at the interface or where the layer that does form is relatively thin. The morphology of the reaction zone, when it is apparent, for the as-fabricated plates is different than what is evident for as-

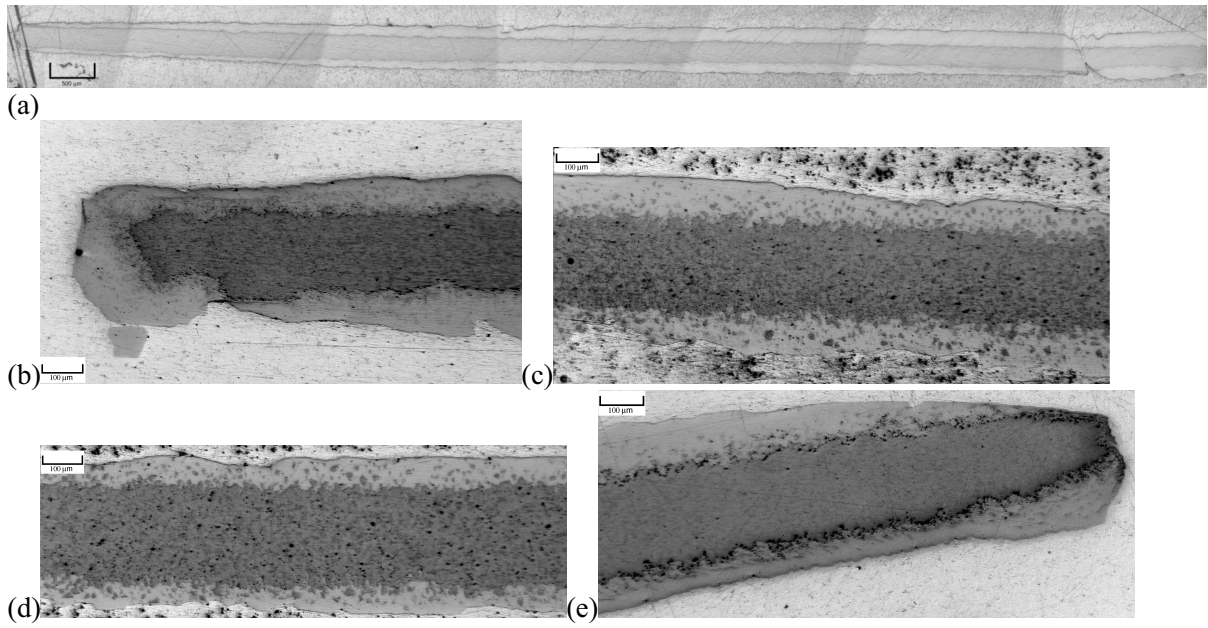


Figure 6. Optical images of (a) a longitudinal cross-section along the bottom 0.75 inch of the fuel plate and (b-e) of different regions along the length of a transverse cross-section of irradiated fuel plate N1F090.

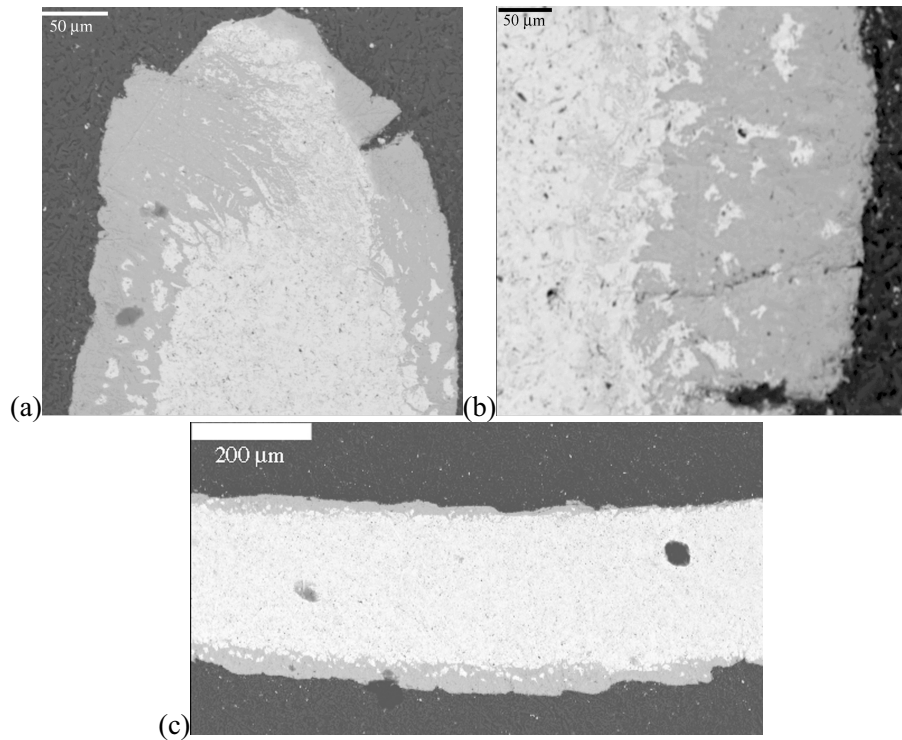


Figure 7. Backscattered electron images of a transverse cross-section taken from as-fabricated plate N1F070.

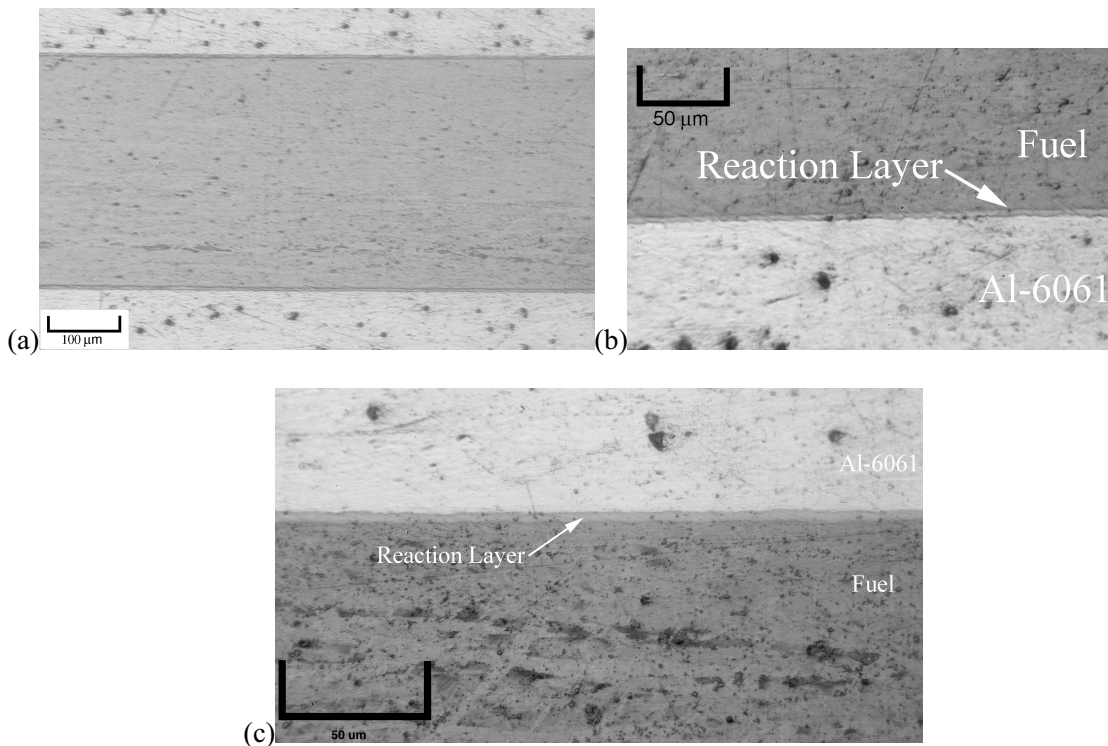


Figure 8. Optical images at various locations along a transverse cross-section taken from fuel plate L1F040.

irradiated plate L1F040. For L1F040, the zone is narrow, uniform and seems to consist of one single layer. Whereas, when the as-fabricated samples display reaction zones, they are comprised of two layers that vary in contrast in an SEM image (see Figures 9a and 9b). Based on the observations that most as-fabricated U-10Mo samples exhibit no reaction and when reaction does occur the formed layer has a morphology different from what is seen for L1F040, there is a reasonable chance that the layer that was observed when conducting PIE on plate L1F040 actually formed during irradiation.

### 3.2.2 L2F030

As was the case for L1F040, the fuel/cladding interface for L2F030 displays a very narrow, uniform layer (see Figure 10). Therefore, this layer also seems to have formed during irradiation. The morphology of this layer is different than what is observed when layers form during fabrication (see Figures 9a and 9b).

### 3.2.3 L1F100

At least based on the cross-sections analyzed to date, L1F100 seems to be the only U-10Mo plate with interaction layers at the fuel/cladding interface that formed during fabrication. These layers are depicted in the optical images taken of a transverse cross-section that are presented in Figure 11. This relatively thick reaction zone has a different morphology than what was observed for L1F040 and L2F030. Both L1F040 and L2F030 have narrow, uniform zones. Whereas, L1F100 has a zone comprised of two layers each with a different morphology. This reaction zone is instead very similar to what was observed for as-fabricated sample L1F080 (see Figures 9a and 9b). This suggests that this layer was formed during fabrication. The compositions of the layers

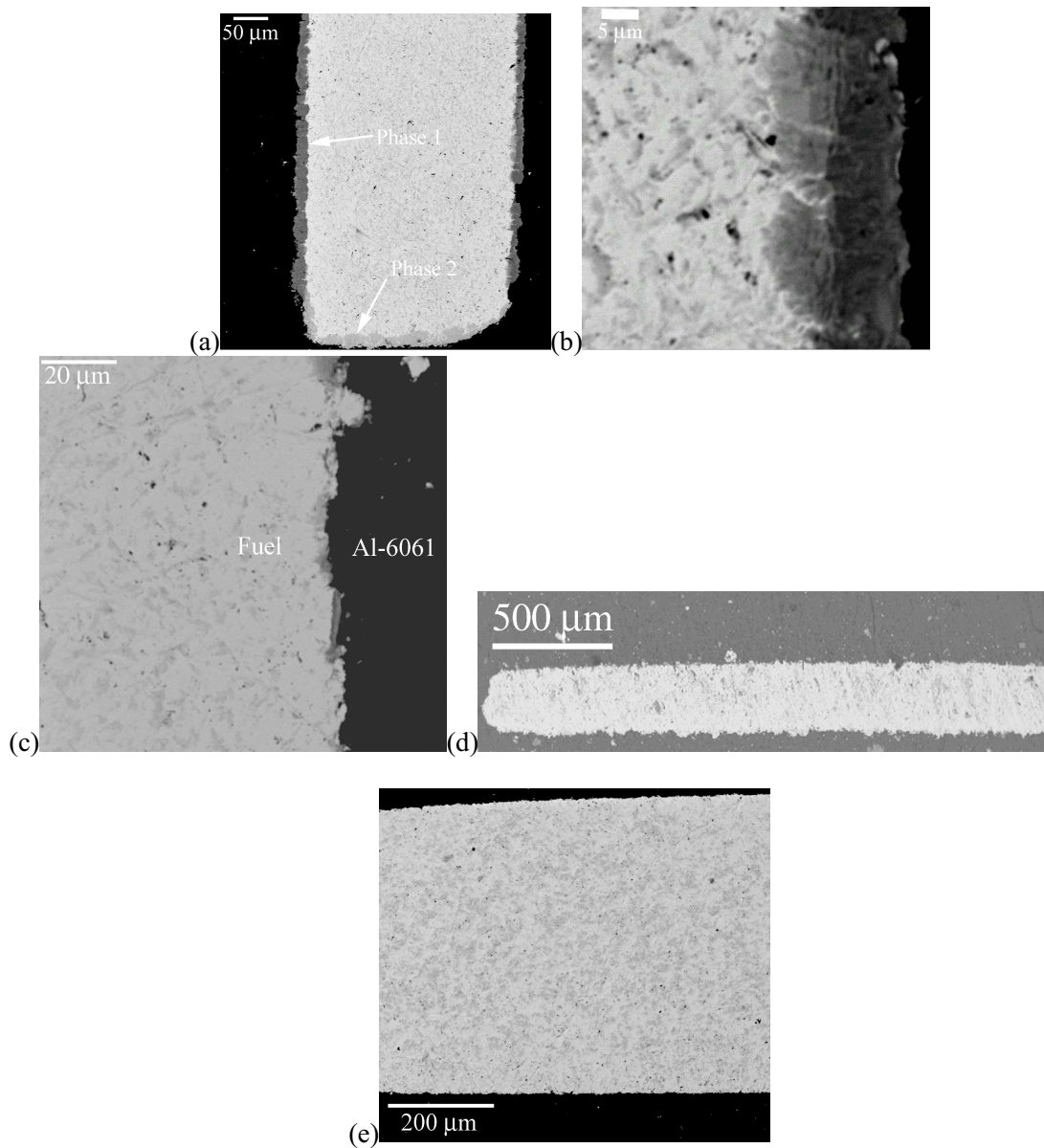


Figure 9. Backscattered electron micrographs of cross-sections from L1F080 (a-c), (d) L1F110, and (e) L2F010.

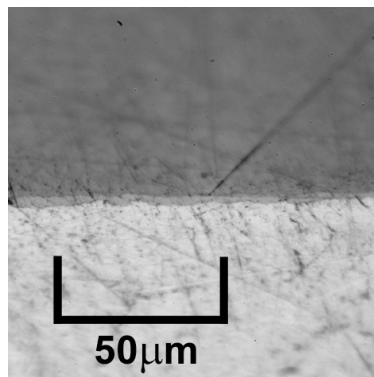


Figure 10. Optical micrograph of the reaction layer observed at the fuel/cladding interface for samples L2F030.



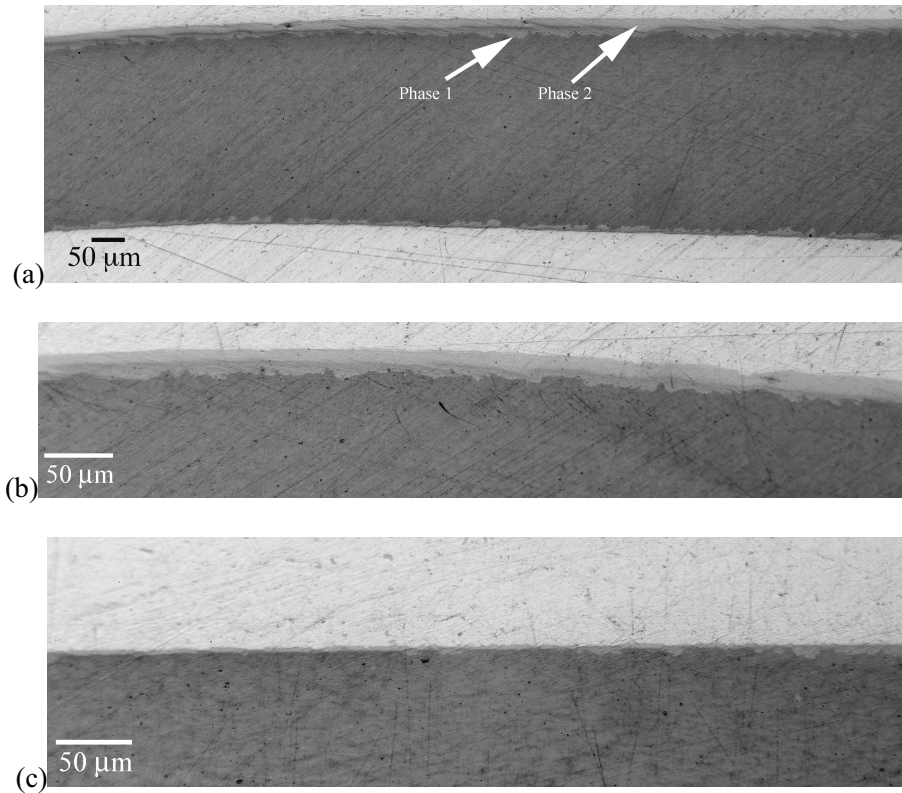


Figure 11. Optical images (a,b) that show areas along a transverse cross section from L1F100 where the fuel/cladding interaction zone is the thickest. (c) is an area adjacent to (a,b) where the interaction zone is relatively narrow. The arrows in (a) identify two distinct phase layers, based on morphology and contrast.

that can form during fabrication of FB monolithic plates have been reported in [4]. Based on these results, phase 1 in Figure 11a was potentially a  $(\text{U},\text{Mo})_{0.9}(\text{Al},\text{Si})_4$  phase with up to 18 at% Si and phase 2 was possibly a  $(\text{U},\text{Mo})(\text{Si},\text{Al})_2$  phase with up to 45 at% Si, before irradiation. In other areas, a different, very narrow reaction zone is observed at the fuel/cladding interface (see Figure 11c). This layer may have developed during irradiation. The largest reaction zones for this plate were observed at the two ends, as shown in Figure 12. This has been commonly observed for as-fabricated fuel plates.

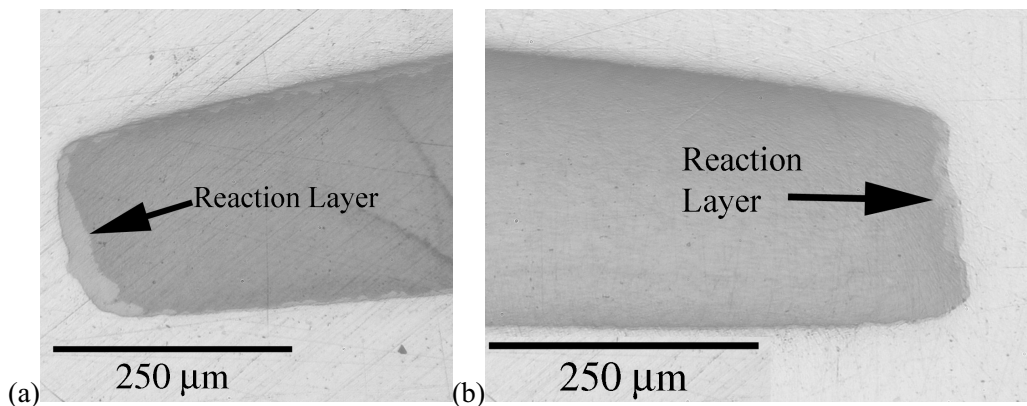


Figure 12. Optical images taken at each end of the L1F100 fuel plate.



#### 4. Discussion

After comparing the results from the PIE of RERTR-6 fuel plates with those from the characterization of after-fabrication sibling samples, a variety of important observations can be made. Firstly, the fuel foils themselves exhibited stable behavior. Even though they were two-phase materials, where one of the phases contained Al, they did not display observable porosity. This shows that some phases that contain U, Mo, and Al will not develop porosity when exposed to the reactor conditions of this experiment. Only N1F030 had (U, Mo, Al)-containing phases that developed porosity at moderate reactor operating conditions, and these phases were present at the fuel/cladding interface, not in the fuel foil itself.

All the plates, except N1F090, seemed to develop new fuel/cladding interaction layers due to the irradiation process, and N1F090 had extensive pre-existing fuel/cladding interaction layers due to fabrication that may have impeded new reactions during irradiation. When layers form at the fuel/cladding interface during irradiation, they exhibit morphologies that are different from those that form during fabrication. This makes them identifiable. Based on the fact that porosity does not seem to form during irradiation in layers present after fabrication, it does not seem necessarily detrimental to a monolithic fuel plate, from a swelling standpoint, if there is some interaction between the fuel and cladding during fabrication.

The composition of the fuel seems to play an important role in determining the overall thickness of the fuel/cladding interaction layer that will develop during fabrication. The U-7Mo plates generally develop larger fuel/cladding interaction zones during fabrication. This has also been demonstrated in annealing studies that have been performed using FB fuel plates [4,5]. Yet, during irradiation, both U-7Mo and U-10Mo monolithic fuel plates appear to form narrow, uniform layers.

Since it is not clear how much Si is in the layers that form due to irradiation (to date, no composition analysis in an SEM has been performed on irradiated monolithic samples), it is not obvious how the presence of Si affects porosity development in interaction zones that develop at the fuel/cladding interface during irradiation in monolithic fuel plates. Yet, for the plates that had pre-existing fuel/cladding interaction layers due to fabrication, it is known that some layers had Si and others did not, and none of these layers developed porosity. Only N1F030 developed porosity at the fuel/cladding interface and this was in a layer that appears to have developed during irradiation. Lastly, since having interaction layers present at the fuel/cladding interface can impact fuel performance in terms of affecting the tenacity of the bond at the fuel/cladding interface (as shown by the debonding of some samples during sample polishing), it is of interest to mitigate the development of these phases. Therefore, the time that monolithic plates are exposed to higher temperatures should be limited, and so the “flattening” and “homogenization anneal” steps should be eliminated from the fuel fabrication process.

Other factors need to be considered when investigating how porosity develops in interaction layers in RERTR fuel plates. It has been proposed that the fission density, temperature, fission rate, interaction layer thickness, and composition that exist at the interface between the fuel and cladding will influence whether or not porosity develops in growing interaction product. This has been suggested for RERTR dispersion fuels [6]. In two cross-sections taken from N1F030, porosity was observed at the fuel/cladding interface (which shows the reproducibility of the observation that porosity is present in the interaction product in this particular fuel plate). The presence of porosity suggests that this fuel plate has exceeded some particular reactor condition or combination of reactor conditions whereby porosity will develop. On the other hand, the other monolithic fuel plates may not have exceeded such condition(s), since porosity was not observed in any of the samples characterized to date. Yet, the reactor conditions for all the fuel plates were

very similar. So, if N1F030 developed porosity all the plates should have developed porosity. Therefore, porosity development must also be dependent on some other factor(s). There may also be a dependency on the initial state of a fuel plate (e.g., alloy composition, foil microstructure, fabrication parameters employed, etc.), which when combined with some specific reactor conditions will result in porosity development.

It has been observed that the interaction product that forms during irradiation is most likely amorphous, at least in dispersion fuels [7]. If this is also the case for monolithic fuels, then this sets this reaction product apart from what forms due to the thermal diffusion that takes place during fabrication. X-ray diffraction analyses of reaction products that form due to thermal diffusion in FB monolithic plates have shown that these products are crystalline [8]. Therefore, the crystalline phases forming during fabrication seem to be more resistant to swelling than are the phases that form due to radiation enhanced diffusion. This agrees with the irradiation behaviors of dispersion fuels comprised of  $\text{UAl}_3$  or  $\text{UAl}_4$  fuel particles that exhibit stable behavior [9]. These uranium-aluminides have U/Al concentration ratios that are similar to those exhibited by the types of phases that form during thermal diffusion in the current RERTR monolithic fuels [4,5]. For the layers that form during the irradiation of RERTR-6 monolithic plates, it has been observed that the majority of the layers did not form porosity. This suggests that if the phases in these layers do go amorphous during irradiation it is not a given that porosity will develop in the phase.

RERTR-7A was the next experiment that tested FB monolithic plates, at even more aggressive reactor conditions [1]. It subjected U-10Mo plates to higher power (surface heat flux  $\sim 215\text{-}325\text{ W/cm}^2$ ), higher temperature (centerline temperature at BOL  $\sim 120\text{-}165^\circ\text{C}$ ) and higher burnup ( $\sim 60\text{-}90\%$  LEU) than the plates in the RERTR-6 experiment. Porosity was observed to develop in the interaction zones of the two U-10Mo RERTR-7A plates that have been destructively examined [10]. This porosity was very similar to what is shown in Figs. 4c and 4d. The plates in RERTR-7A did not go through the flattening and homogenization anneal steps that were performed on the plates that went into RERTR-6, which means that the amount of pre-existing interaction layers, due to fabrication, was negligible. Therefore, any layers that were present at the fuel/cladding interface formed during irradiation. By comparing the porosity that formed for the RERTR-7A plates with what was observed for the irradiated RERTR-6 plates, it appears that the reactor conditions for the RERTR-6 experiment were just at the edge of what is required for causing porosity to develop in irradiation-induced, fuel/cladding interaction layers in U-7Mo and U-10Mo monolithic fuels, and the RERTR-7A experiment was well into the regime where porosity will develop in interaction layers that form during irradiation.

## **5. Conclusions**

Based on the comparison of results from the SEM characterization of as-fabricated RERTR-6 monolithic fuel plates with those from the characterization of irradiated RERTR-6 fuel plates using optical metallography, it can be concluded that (1) fuel/cladding interaction layers formed during fabrication exhibit good irradiation performance under reactor conditions like those of the RERTR-6 experiment, (2) fuel foils that have Al-containing phases present in the alloy microstructure also exhibit favorable irradiation performance, (3) the morphology of layers that form during fabrication is different from those that form during irradiation and therefore can each be identified using optical metallography, (4) and due to the appearance of porosity in the irradiation-induced interaction layers for some RERTR-6 monolithic samples but not for others, the reactor conditions of the RERTR-6 experiment were just severe enough to induce the formation of such pores.

## Acknowledgments

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